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Supplementary webappendix

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Supplement to: Smith KR, Jerrett M, Anderson HR, et al. Public health benefits of strategies to reduce greenhouse-gas emissions: health implications of short-lived greenhouse pollutants. *Lancet* 2009; published online Nov 25. DOI:10.1016/S0140-6736(09)61716-5.

Webappendix
Public Health Benefits of Strategies to Reduce Greenhouse-Gas Emissions: Health Implications of the Short-Lived Greenhouse Pollutants

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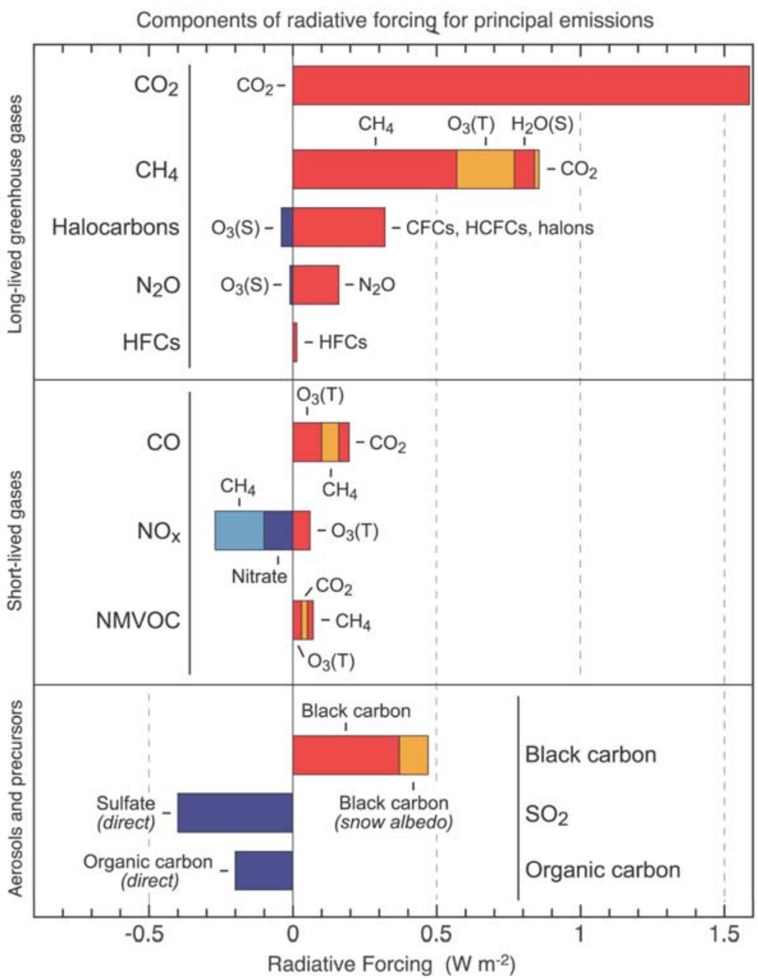
I. Box Figure 1. Radiative Forcing (RF) in 2005 due to human emissions since 1750.



Box Figure 1. Radiative Forcing (RF) in 2005 due to human emissions since 1750. This graph is focused on the emitted species and thus shows both the direct and indirect climate impacts that result for each. Several affect ozone levels, for example, but ozone itself is not emitted directly. Some, such as NO_x, have both warming and cooling (negative RF) impacts. Others, like methane, have both direct and indirect impacts. Several are eventually oxidized to CO_2 in the atmosphere and, if from fossil sources, contribute to net warming by this route as well. Based on Forster et al. (2007)[1]

1. Forster, P., V. Ramaswamy, P. Artaxo, T. Berntsen, R. Betts, D.W. Fahey, J. Haywood, J. Lean, D.C. Lowe, G. Myhre, J. Nganga, R. Prinn, G. Raga, M. Schulz and R. Van Dorland, 2007: Changes in Atmospheric Constituents and in Radiative Forcing. Chapter 2 in*: Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change* [Solomon, S., D. Qin, M. Manning, Z. Chen, M. Marquis, K.B. Averyt, M.Tignor and H.L. Miller (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.

II. Primer on Black Smoke, Black Carbon, and Elemental Carbon Metrics

Summary

Black carbon (BC) is the term used by the climate-modeling community to describe small soot particles that are highly warming per unit mass. As yet, there is only limited monitoring data of BC for epidemiologic analyses. Black smoke (BS) measurements are closely related, date back to the 1920s, and continue to be collected today. BS data provide the largest evidence base for epidemiologic studies of a measure related to BC. It is not straightforward, however, to convert one to the other in a quantitative way across studies [1, 2]. Elemental carbon (EC), determined by a thermal-chemical technique, has a more straightforward relationship with BC and has been measured in a few air pollution monitoring networks.

Measurement methods

The Black Smoke Index

BS measurements, reported in units of μg m^{-3}, are taken by pumping a known volume of air through a known area of filter paper, and then measuring the darkness of the stain by its reflectance of white light[2]. The reflectance is measured relative to the light reflected by a clean filter of the same material. These measurements may be made using an instrument such as the EEL M43D Smokestain Reflectometer where the reflectance, volume, and area are converted to a BS Index using a standard table.[2] The OECD convention for BS is derived from the following equation:

$$I_{\mathrm{BS}}\,(\mu\mathrm{g\,m}^{-3}) = 8.655 \times 10^{8}\alpha^{2} + 2.219 \times 10^{5}\alpha \qquad \text{(eq.1)}^{2}$$

where I_{BS} is the BS Index and α is the absorption coefficient of the sampled air, in units of m^{-1}. The British Smoke Stain convention for BS is equal to the OECD version multiplied by a factor of 0.8667. For a complete discussion of the derivation of the Black Smoke Index, please refer to Quincey et al. (2007).[2]

Black Carbon

The measurement of BC is performed by an optical <u>transmission</u> method, as distinct from the optical <u>reflectance</u> method for BS. The sample is collected on a filter and the optical transmission is measured either in 'batch mode' for a time-weighted average determination; or incrementally while the filter is collecting, to provide real-time data. This may be performed at multiple optical wavelengths to provide additional information or speciation. Following Quincey (2007)[2], the BC concentration can be determined by the following equation:

$$C_{\mathrm{BC}}\,(\mu\mathrm{g\,m}^{-3}) = 10^{6}\alpha\,(\mathrm{m}^{-1})/\alpha_{\mathrm{atn}}\,(\mathrm{m}^{2}\,\mathrm{g}^{-1}). \qquad \text{(eq. 2)}^{2}$$

where C_{BC} is the BC concentration; α is the absorption coefficient; and σ_{atn} is the mass extinction coefficient for particles collected on the filter. This, in practice, depends on the particle size, particle composition, wavelength of light used, and filter type; and is determined empirically.[2] Quincey (2007) uses a σ_{atn} value of 16.6 m^2g^{-1} taken from the Magee AE21 Aethalometer instrument literature. From this,

$$C_{\mathrm{BC}}\,(\mu\mathrm{g\,m}^{-3}) = 6.02 \times 10^{4}\alpha \qquad \text{(eq. 3)}^{2}$$

For a complete discussion of the derivation of the above equation, refer to Quincey (2007).[2]

Elemental Carbon

The material denoted "Elemental" Carbon (EC) is determined by thermal-optical analysis in a multi-step process. The particulate matter (PM) is usually collected on quartz fiber filters and then analyzed at different temperatures in inert or oxidizing atmospheres. Carbon released from the sample is converted to CO_2 or methane and detected during the progressive analysis. Assignment of the precise fraction of the analyte to "EC" is still an active topic of research[3,4], and there are several different protocols, see Environmental Protection Agency (2003)[5]. These include the National Institute of Occupational Safety and Health (NIOSH) method 5040; the "IMPROVE-A" protocol developed by the Desert Research Institute, NV; and the "EUSAAR" protocol of JRC, Ispra (Europe). In this paper, we used EC data from ambient air PM samples analyzed by the NIOSH method.

Because there are many techniques that have been employed to estimate EC concentrations, a number of studies have examined inter-method comparability. As an example, Miller et al. (2007)[6] compared the NIOSH method with the Thermo Electron Personal Data RAM 1200 photometer (PDR) technique (particulate mass concentration based on light scattering) and found that they agreed well (R^2=0.97) when the data were adjusted for relative humidity (Figure 1).[6]

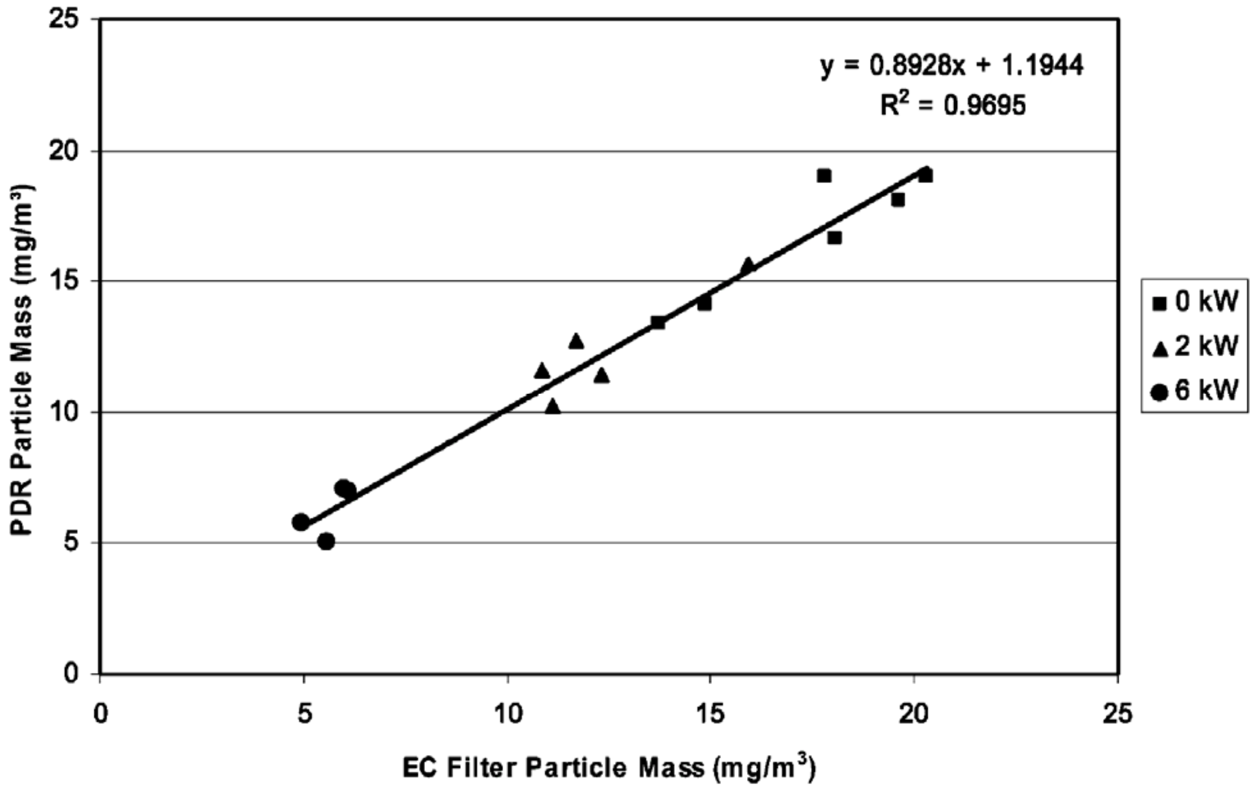


Figure 1: Linear regression of corrected PDR data and EC filter (NIOSH) sample data. Adapted From Miller et al. (2007).[6]

<u>Translation between metrics</u>

Black Smoke to Black Carbon

Because both BS and BC methods essentially measure the same thing – the optical absorption coefficient of airborne particles collected on a filter – Quincey (2007) has shown that a simple, quadratic equation can effectively convert black smoke measurements into black carbon concentration estimates[2]:

$$I_{\mathrm{BS}} = 0.239 C_{\mathrm{BC}}^2 + 3.69 C_{\mathrm{BC}} \qquad \text{(eq. 4)}^2$$

or, conversely,

$$C_{BC} = \sqrt{4.18 I_{BS} + 59.6} - 7.72 \quad \text{(eq. 5)}^2$$

From another, albeit more simple methodological angle, Novakov and Hansen (2004)[7] follow Erdman et al. (1993)[8] and convert their BS data, in units of μg (BS) m^{-3}, to BC data in units of μg (BC) m^{-3} via the linear relationship, BC = 0.23 BS.

The relationships between the BS index and BC (μg m^{-3}) are robust as shown by the tight scatters (Figure 2) from the modeled data from Quincey et al. (2007); and Figure 4 shows this relationship with high R^2 values (urban=0.85; rural=0.75) for empirical data from the Netherlands.[9]

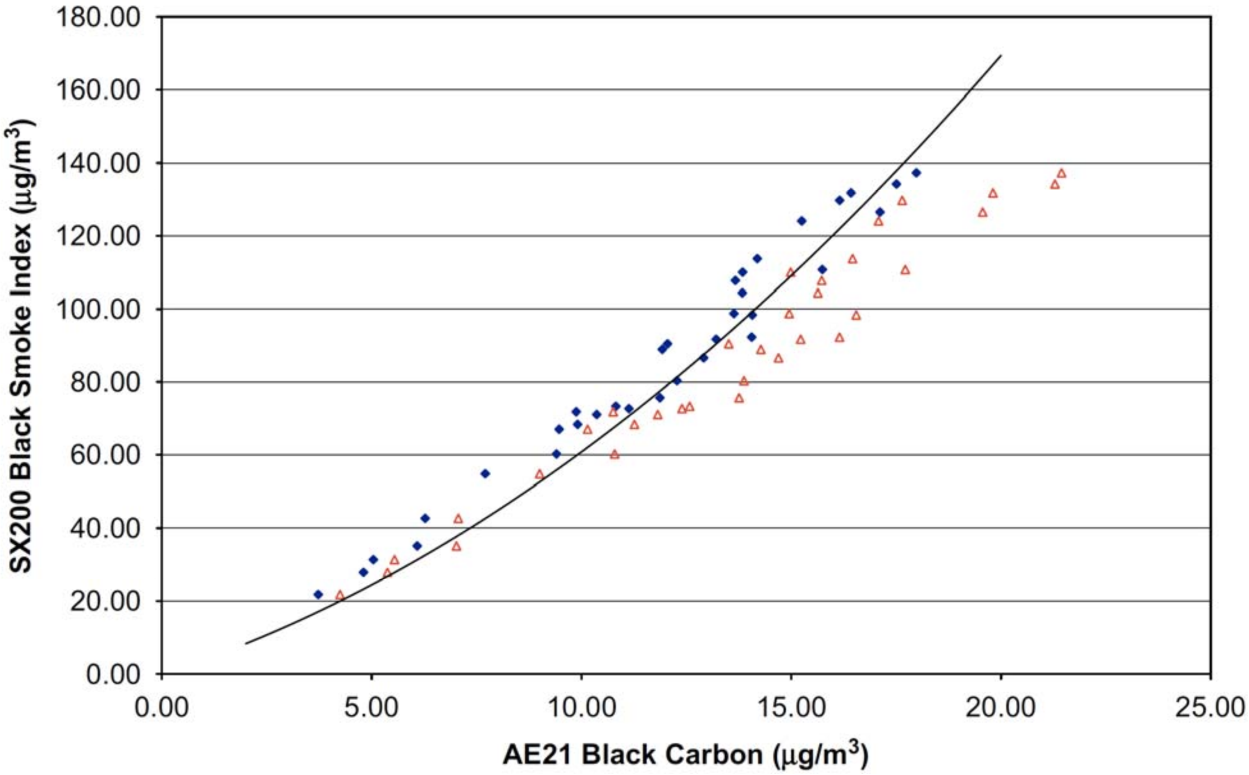


Figure 2: Unprocessed Black Carbon data (dark diamonds) and processed Black Carbon data (light triangles) plotted against the SX200 Black Smoke data, together with the derived theoretical relationship between Black Carbon and Black Smoke measurements (Eq. (4) – black line). From Quincey et al. (2007)[2]

Elemental Carbon to Black Carbon

Although subject to the measurement complexities described above, there is a high correlation between EC and BC (R^2=0.89) (Figure 3).[12] Both optical and thermal methods can account for, or separate, the contributions from 'graphitic' carbon or 'tarry' organic compounds[2]: optical methods can analyze at multiple wavelengths while thermal methods can employ different temperature profiles or oxidizing conditions. EC is interchangeable with BC insofar as both of the components, regardless of their physiochemical definition, represent the vast majority of light-absorbing components in undifferentiated PM and are thus significant warming agents.[11] Because of this, many researchers struggle with criteria that can distinguish between the two and often conflate the terms.[13]

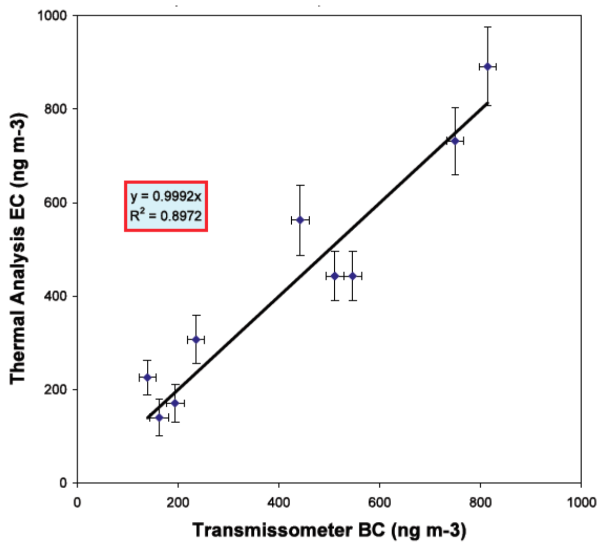


Figure 3: Comparison of Optical Transmission (BC) measurements with Thermal (EC) Analysis. Data Source: Ahmeda et al. (2009).[12]

Moreover, Schaap et al. (2007)[9] notes that the relation between BS and EC is linear and highly correlated, albeit dependent on the type of location (Figure 4). The authors propose that EC can be predicted using the following relationships for rural and urban locations:

$$\mathrm{EC} = \begin{cases} 0.056 \times \mathrm{BS} + 0.16 & \text{(rural)}, \\ 0.088 \times \mathrm{BS} + 0.32 & \text{(urban)}. \end{cases}$$

(eq. 6) Cited from: Schaap et al. (2007)[9]

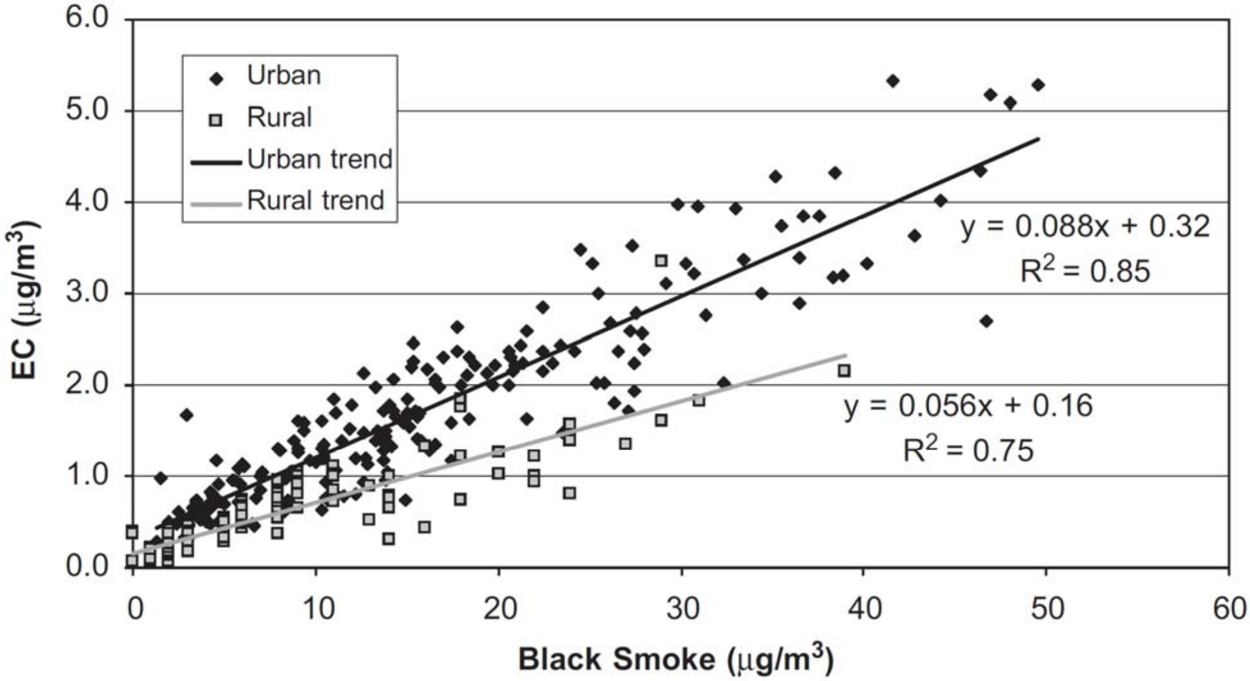


Figure 4: Elemental carbon as a function of Black Smoke at rural and urban locations. Cited from: Schaap et al. (2007)[9]

There are a wide range of inter-species delineations between EC and BC, as well as intra-species differences between various physiochemical forms of BC. Both EC and BC data can be used for environmental epidemiology for the following reasons:

1. EC and BC data both represent emissions from combustion processes. Neither represents a unique chemical formulation.
2. Both BC and EC data metrics indicate the mass of nearly all the light-absorbing constituents of undifferentiated PM.[11]
3. Carbonaceous particles of combustion origin tend to be diametrically small[14-18]. PM of diesel origin tends to be finer than that from other sources such as coal or biomass combustion.[19] Ultrafine particles, however, tend to agglomerate soon after release from the emission source[20], and BC is usually found in the ambient atmosphere in the size ranges of 0.1 to 0.5 µm. Nonetheless, it is thought that EC and BC may have effects both chemically and as a generic function of being particles (see main paper for further discussion).
4. These carbonaceous particles are nearly always associated with other – often toxic – constituents such as polycyclic aromatic hydrocarbons (PAHs), and other organic compounds.[20] The California Air Resources Board (CARB) uses BC as a surrogate for Diesel Particulate Matter (DPM), which has been designated as a Toxic Air Contaminant (TAC)[21]

As the cohort we examine in the main paper is all urban and in one country, we consider that EC is as good an indicator of what climate scientists refer to as BC as can currently be found in air pollution measurement networks.

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III. Supplemental material for Time Series Systematic Reviews

Methods

The Air Pollution Epidemiology Database (APED), a database of results from time series studies of the health effects of outdoor air pollution, provided city specific standardized effect estimates for sulphate (SO_4^{2-}) and BS. Details of APED have been published elsewhere[1,2]. In brief, the database comprises a bibliography of time series studies published up to May 2009 and a relational database of effect estimates and associated information extracted from selected studies. Published, peer reviewed time series studies were identified through systematic searching of Medline, Embase, and Web of Science databases with no constraints placed on language or date of publication. The search criteria were developed using known papers and then tested several times against citations in published reviews of the literature. A two stage sifting and assessment process was then applied to identify daily time series studies providing the necessary data for quantitative analysis. This assessment considered the appropriateness of the statistical methods used, data quality and the provision of the information needed to standardize the effect estimates. The data characterizing the effect estimates were entered into a relational database (ACCESS, Microsoft Corp.) where routines to calculate standardized estimates were applied. Studies reporting associations between all cause, cardiovascular and respiratory mortality and SO_4^{-2}, BS and O_3 were identified and their results extracted into STATA. City specific results were included in a meta-analysis if the estimate was accompanied by an indication of the precision of the estimate. When more than one estimate for a specific location was available only the most recently published estimate for that location was included in the meta-analysis. Summary fixed and random effects estimates were calculated using the method of Der Simonian and Laird[3] and implemented in the *metan* procedure in STATA[4]. Results from multi- city studies were also extracted and although not included in the calculation of summary estimates their inclusion in this review was warranted because of the additional evidence they provide.

Sources Cited: Supplemental material for time series systematic reviews – Methods

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Table 1 City specific, fixed and random effects summary estimates and multi-city estimates for sulphate and all cause, all age mortality

Single city/site estimates

Author	Year	Country	City	Conc*	%~	95% CI$
Dockery	1992	USA	St. Louis	8.7	0.61	(-0.52, 1.75)
Dockery	1992	USA	Tennessee eastern	8.0	0.80	(-1.54, 3.20)
Burnett	1998	Canada	Toronto	8.2	0.18	(0.08, 0.27)
Gwynn	2000	USA	Buffalo	5.9	0.23	(0.05, 0.41)
Klemm	2000	USA	Georgia	4.4	0.51	(-0.54, 1.57)
Hoek	2000	Netherlands	Netherlands	3.8	0.13	(0.02, 0.23)
Lippmann	2000	USA	Wayne County	3.2	0.15	(-0.12, 0.42)
Goldberg	2001	Canada	Montreal	2.2	0.51	(0.27, 0.76)
Anderson	2001	UK	West Midlands	2.7	-0.07	(-0.40, 0.26)
Maynard	2007	USA	Boston	2.4	0.49	(0.00, 0.97)
			Pooled FE		*0.18*	*(0.12, 0.24)*
			Pooled RE		*0.21*	*(0.11, 0.30)*
			Heterogeneity chi-squared = 13.55 (d.f. = 9) p = 0.139			

Multi-city estimates

Author	Year	Location	City	%~	95% CI$
Schwartz	1996	USA	6 communities	0.22	(0.13, 0.31)
Brook	2007	Canada	10 cities	0.40	(0.10, 0.70)
Ostro	2007	USA	6 California counties	0.12	(-0.76, 1.00)

* Mean/median daily concentrations (µg/m3)
~ Percentage change in mean number of deaths per 1µg/m3 increase in SO_4^{-2}
$ 95% confidence limits

Table 2 City specific, fixed and random effects summary estimates and multi-city estimates for sulphate and all cardiovascular/cardiac, all age mortality

Single city estimates

Author	Year	Country	City	Conc*	%~	95%CI~
Gwynn	2000	USA	Buffalo	5.9	0.26	(-0.03, 0.55)
Hoek	2000	Netherlands	Netherlands	3.8	0.08	(-0.08, 0.24)
Lippmann	2000	USA	Wayne County	3.2	0.11	(-0.29, 0.52)
Anderson	2001	UK	West Midlands	2.7	-0.23	(-0.70, 0.25)
Maynard	2007	USA	Boston	2.4	-0.09	(-0.67, 0.49)
			Pooled FE		*0.09*	*(-0.04, 0.21)*
			Pooled RE		*0.09*	*(-0.04, 0.21)*

Heterogeneity chi-squared = 3.32 (d.f. = 4) p = 0.506

Multi-city estimates

Author	Year	Location	City	%~	95%CI~
Ostro	2007	USA	6 California counties	0.36	(-0.91, 1.65)

* Mean/median daily concentrations (μg/m3)
~ Percentage change in mean number of deaths per 1μg/m3 increase in SO42-
$ 95% confidence limits

Table 3 City specific, fixed and random effects summary estimates and multi-city estimates for sulphate and all respiratory, all age mortality

Single city estimates

Author	Year	Country	City	Conc*	%~	95% CI$
Gwynn	2000	USA	Buffalo	5.9	0.68	(0.05, 1.31)
Lippmann	2000	USA	Wayne County	3.2	0.40	(-0.61, 1.43)
Anderson	2001	UK	West Midlands	2.7	-0.31	(-1.13, 0.52)
Maynard	2007	USA	Boston	2.4	0.92	(-0.49, 2.36)
			Pooled FE		*0.39*	*(-0.04, 0.82)*
			Pooled RE		*0.37*	*(-0.15, 0.90)*

Heterogeneity chi-squared = 4.11 (d.f. = 3) p = 0.250

Multi-city estimates

Author	Year	Location	City	%~	95% CI$
Ostro	2007	USA	6 California counties	0.70	(-1.65, 3.11)

* Mean/median daily concentrations (μg/m3)
~ Percentage change in mean number of deaths per 1μg/m3 increase in SO42-
$ 95% confidence limits

Table 4 City specific, fixed and random effects summary estimates and multi-city estimates for Black Smoke and all cause, all age mortality

Single city estimates

Author	Year	Country	City	Conc*	%~	95% CI$
Bellido Blasco	1999	Spain	Castellon	20.3	0.15	(-0.05, 0.35)
Aguinaga	1999	Spain	Pamplona	21.7	0.29	(-0.19, 0.78)
Perez	1999	Spain	Vitoria-Gasteiz	45.0	0.06	(-0.03, 0.16)
Katsouyanni	2001	Greece	Athens	64.0	0.07	(0.04, 0.09)
Katsouyanni	2001	Spain	Barcelona	39.0	0.16	(0.10, 0.21)
Katsouyanni	2001	Spain	Bilbao	23.0	0.08	(-0.07, 0.23)
Katsouyanni	2001	UK	Birmingham	11.0	0.03	(-0.06, 0.13)
Katsouyanni	2001	Poland	Cracow	36.0	-0.02	(-0.06, 0.02)
Katsouyanni	2001	Slovenia	Ljubljana	13.0	-0.01	(-0.13, 0.11)
Katsouyanni	2001	Poland	Lodz	30.0	-0.01	(-0.05, 0.04)
Katsouyanni	2001	Poland	Poznan	23.0	0.06	(0.02, 0.11)
Saez	2001	Spain	Vigo	102.6	0.07	(0.01, 0.13)
Anderson	2001	UK	West Midlands	10.9	0.04	(-0.09, 0.16)
Katsouyanni	2001	Poland	Wroclaw	33.0	0.03	(-0.02, 0.07)
Arribas-Monzon	2001	Spain	Zaragoza	n/a	0.04	(-0.04, 0.11)
Le Tertre	2002	France	Bordeaux	13.2	0.15	(0.00, 0.30)
Le Tertre	2002	France	Le Havre	n/a	0.02	(-0.15, 0.20)
Le Tertre	2002	France	Marseille	16.0	0.12	(0.05, 0.19)
Le Tertre	2002	France	Paris	16.0	0.04	(0.00, 0.08)
Le Tertre	2002	France	Rouen	14.1	0.01	(-0.10, 0.13)
Ballester	2002	Spain	Valencia	44.2	0.18	(0.09, 0.27)
Pattenden	2003	UK	London	n/a	0.08	(-0.67, 0.84)
Goodman	2004	Ireland	Dublin	40.0	0.04	(0.03, 0.05)
Bogdanovic	2006	Serbia	Nis	23.0	0.11	(0.01, 0.22)
Fischer	2009	Netherlands	Netherlands	6.0	-0.03	(-0.10, 0.04)

Pooled FE	*0.04*	*(0.04, 0.05)*
Pooled RE	*0.05*	*(0.03, 0.07)*

Heterogeneity chi-squared = 62.09 (d.f. = 24) p = 0.000

Multi-city estimates

Author	Year	Location	City	%~	95% CI$
Katsouyanni	1997	Europe	APHEA 1	0.03	(0.02, 0.03)
Aga	2003	Europe	APHEA 2 - 29 cities	0.06	(0.03, 0.08)
Ballester	2003	Spain	13 cities	0.08	(0.04, 0.12)
Carder	2008	Scotland	9 sites	0.17	(0.07, 0.26)

* Mean/median daily concentrations (μg/m3)
~ Percentage change in mean number of deaths per 1 μg/m3 increase in BS
$ 95% confidence limits

Table 5 City specific, fixed and random effects summary estimates and multi-city estimates for Black Smoke and all cardiovascular/cardiac, all age mortality

Single city estimates

Author	Year	Country	City	Conc*	%~	95% CI$
Wojtyniak	1996	Poland	Krakow	73.3	0.01	(-0.02, 0.05)
Wojtyniak	1996	Poland	Lodz	57.3	0.01	(-0.02, 0.05)
Wojtyniak	1996	Poland	Poznan	34.0	-0.02	(-0.08, 0.04)
Wojtyniak	1996	Poland	Wroclaw	54.3	0.01	(-0.04, 0.06)
Cambra	1999	Spain	Bilbao	23.1	-0.17	(-0.37, 0.04)
Bellido Blasco	1999	Spain	Castellon	20.3	0.34	(0.05, 0.64)
Bremner	1999	UK	London	10.8	0.12	(-0.01, 0.25)
Aguinaga	1999	Spain	Pamplona	21.7	-0.23	(-1.04, 0.58)
Garcia-Aymerich	2000	Spain	Barcelona	42.4	0.11	(0.04, 0.19)
Anderson	2001	UK	West Midlands	10.9	0.09	(-0.09, 0.27)
Arribas-Monzon	2001	Spain	Zaragoza	n/a	0.07	(-0.05, 0.18)
Le Tertre	2002	France	Bordeaux	13.2	0.17	(-0.07, 0.42)
Le Tertre	2002	France	Marseille	16.0	0.09	(-0.02, 0.21)
Le Tertre	2002	France	Paris	16.0	0.04	(-0.02, 0.10)
Le Tertre	2002	France	Rouen	14.1	0.10	(-0.11, 0.30)
Ballester	2002	Spain	Valencia	44.2	0.15	(0.01, 0.29)
Goodman	2004	Ireland	Dublin	40.0	0.04	(0.02, 0.06)
Filleul	2006	France	Le Havre	11.0	0.44	(-0.09, 0.97)
Stankovic	2007	Serbia	Nis	22.8	0.13	(-0.03, 0.29)
Fischer	2009	Netherlands	Netherlands	6.0	-0.12	(-0.24, 0.00)
			Pooled FE		*0.03*	*(0.02, 0.04)*
			Pooled RE		*0.04*	*(0.01, 0.06)*

Heterogeneity chi-squared = 36.82 (d.f. = 19) p = 0.008

Multi-city estimates

Author	Year	Location	City	%~	95% CI$
Analitis	2006	Europe	APHEA 2 - 15 cities	0.06	(0.03, 0.09)
Ballester	2003	Spain	13 cities	0.03	(-0.02, 0.08)
Carder	2008	Scotland	9 sites	0.04	(-0.10, 0.18)

* Mean/median daily concentrations (μg/m3)
~ Percentage change in mean number of deaths per 1 μg/m3 increase in BS
$ 95% confidence limits

Table 6 City specific, fixed and random effects summary estimates and multi-city estimates for Black Smoke and all respiratory, all age mortality

Single city estimates

Author	Year	Country	City	Conc*	%~	95% CI$
Wojtyniak	1996	Poland	Krakow	73.3	-0.02	(-0.14, 0.09)
Wojtyniak	1996	Poland	Lodz	57.3	-0.08	(-0.19, 0.02)
Wojtyniak	1996	Poland	Poznan	34.0	-0.10	(-0.28, 0.08)
Wojtyniak	1996	Poland	Wroclaw	54.3	-0.19	(-0.34, -0.03)
Cambra	1999	Spain	Bilbao	23.1	0.29	(-0.11, 0.70)
Bellido Blasco	1999	Spain	Castellon	20.3	0.36	(-0.26, 0.98)
Bremner	1999	UK	London	10.8	0.19	(0.02, 0.36)
Aguinaga	1999	Spain	Pamplona	21.7	1.26	(-0.39, 2.94)
Garcia-Aymerich	2000	Spain	Barcelona	42.4	0.10	(-0.05, 0.25)
Anderson	2001	UK	West Midlands	10.9	0.01	(-0.29, 0.31)
Arribas-Monzon	2001	Spain	Zaragoza	n/a	0.29	(0.06, 0.51)
Le Tertre	2002	France	Bordeaux	13.2	0.20	(-0.37, 0.77)
Le Tertre	2002	France	Marseille	16.0	0.26	(0.02, 0.50)
Le Tertre	2002	France	Paris	16.0	-0.02	(-0.15, 0.11)
Le Tertre	2002	France	Rouen	14.1	0.07	(-0.33, 0.46)
Ballester	2002	Spain	Valencia	44.2	-0.21	(-0.50, 0.08)
Goodman	2004	Ireland	Dublin	40.0	0.09	(0.05, 0.13)
Filleul	2006	France	Le Havre	11.0	0.45	(-0.64, 1.55)
Bogdanovic	2006	Serbia	Nis	23.0	0.17	(-0.09, 0.44)
Fischer	2009	Netherlands	Netherlands	6.0	0.03	(-0.19, 0.25)

Pooled FE	*0.06*	*(0.03, 0.09)*
Pooled RE	*0.04*	*(-0.02, 0.11)*

Heterogeneity chi-squared = 43.88 (d.f. = 19) p = 0.001

Multi-city estimates

Author	Year	Location	City	%~	95% CI$
Analitis	2006	Europe	APHEA 2 - 15 cities	0.08	(0.01, 0.16)
Ballester	2003	Spain	13 cities	0.11	(0.04, 0.18)
Carder	2008	Scotland	9 sites	0.52	(0.29, 0.76)

* Mean/median daily concentrations (ug/m3)
~ Percentage change in mean number of deaths per 1 μg/m3 increase in BS
$ 95% confidence limits

Table 7 City specific, fixed and random effects summary estimates and multi-city estimates for ozone (8 hour) and all cause, all age mortality

Single city estimates

Author	Year	Country	City	Conc*	%~	95% CI$
Simpson	1997	Australia	Brisbane	16.7	0.12	(0.04, 0.20)
Borja-Aburto	1997	Mexico	Mexico City	94.0	0.02	(0.01, 0.03)
Michelozzi	1998	Italy	Rome	21.0	0.04	(0.00, 0.08)
Hong	1999	South Korea	Inchon	15.4	-0.02	(-0.05, 0.00)
Bremner	1999	UK	London	16.0	-0.01	(-0.05, 0.02)
Cadum	1999	Italy	Turin	73.7	0.03	(-0.01, 0.08)
Klemm	2000	USA	Georgia	40.6	-0.02	(-0.13, 0.09)
Hoek	2000	Netherlands	Netherlands	47.0	0.02	(0.01, 0.03)
Wong	2001	China	Hong Kong	33.5	0.02	(-0.02, 0.05)
Anderson	2001	UK	West Midlands	24.0	0.05	(0.00, 0.10)
Saez	2002	Spain	Barcelona	67.5	0.01	(-0.02, 0.04)
Le Tertre	2002	France	Le Havre	43.4	0.07	(-0.05, 0.18)
Le Tertre	2002	France	Lyon	52.0	0.07	(0.00, 0.15)
Saez	2002	Spain	Madrid	42.1	0.03	(-0.01, 0.06)
Le Tertre	2002	France	Paris	26.0	0.04	(0.00, 0.08)
Le Tertre	2002	France	Rouen	57.9	0.10	(0.00, 0.21)

Le Tertre	2002	France	Strasbourg	37.0	0.06	(-0.01, 0.12)
Le Tertre	2002	France	Toulouse	68.0	0.03	(-0.08, 0.14)
Saez	2002	Spain	Valencia	45.5	0.22	(0.05, 0.40)
Parodi	2005	Italy	Genoa	79.2	0.06	(0.01, 0.11)
Zhang	2006	China	Shanghai	56.1	0.04	(0.02, 0.07)
Qian	2007	China	Wuhan	47.2	0.02	(-0.01, 0.05)

Pooled FE	*0.02*	*(0.02, 0.03)*
Pooled RE	*0.03*	*(0.02, 0.04)*

Heterogeneity chi-squared = 47.55 (d.f. = 21) p = 0.001

Multi-city estimates

Author	Year	Location	City	Conc*	%~	95% CI$
Gryparis	2004	Europe	23 cities	n/a	0.003	(-0.018, 0.024)

* Mean/median daily concentrations (ug/m3)
~ Percentage change in mean number of deaths per 1 ug/m3 increase in O3
$ 95% confidence limits

Table 8 City specific, fixed and random effects summary estimates and multi-city estimates for ozone (8 hour) and cardiovascular, all age mortality

Single city estimates

Author	Year	Country	City	Conc*	%~	95% CI$
Borja-Aburto	1997	Mexico	Mexico City	94.0	0.04	(0.02, 0.06)
Hong	1999	South Korea	Inchon	15.4	-0.09	(-0.65, 0.47)
Bremner	1999	UK	London	16.0	0.07	(0.01, 0.12)
Cadum	1999	Italy	Turin	73.7	0.07	(0.00, 0.14)
Hoek	2001	Netherlands	Netherlands	n/a	0.04	(0.02, 0.05)
Anderson	2001	UK	West Midlands	24.0	0.02	(-0.06, 0.09)
Saez	2002	Spain	Barcelona	67.5	0.06	(0.00, 0.11)

Wong	2002	China	Hong Kong	29.3	-0.03	(-0.09, 0.03)
Le Tertre	2002	France	Le Havre	43.4	0.03	(-0.18, 0.24)
Le Tertre	2002	France	Lyon	52.0	0.04	(-0.16, 0.24)
Saez	2002	Spain	Madrid	42.1	0.05	(0.00, 0.11)
Le Tertre	2002	France	Paris	26.0	0.04	(-0.03, 0.12)
Le Tertre	2002	France	Rouen	57.9	0.14	(-0.06, 0.34)
Le Tertre	2002	France	Strasbourg	37.0	0.02	(-0.08, 0.13)
Le Tertre	2002	France	Toulouse	68.0	0.10	(-0.09, 0.29)
Saez	2002	Spain	Valencia	45.5	0.31	(0.04, 0.58)
Penttinen	2004	Finland	Helsinki	50.0	0.04	(-0.05, 0.12)
Zhang	2006	China	Shanghai	56.1	0.05	(0.01, 0.10)
Qian	2007	China	Wuhan	47.2	0.02	(-0.05, 0.10)

Pooled FE	*0.04*	*(0.03, 0.05)*
Pooled RE	*0.04*	*(0.03, 0.05)*

Heterogeneity chi-squared = 13.61 (d.f. = 18) p = 0.754

* Mean/median daily concentrations (ug/m3)
~ Percentage change in mean number of deaths per 1 ug/m3 increase in O3
$ 95% confidence limits

Table 9 City specific, fixed and random effects summary estimates and multi-city estimates for ozone (8 hour) and respiratory, all age mortality

Single city estimates

Author	Year	Country	City	Conc*	%~	95% CI$
Simpson	1997	Australia	Brisbane	16.7	0.19	(-0.09, 0.48)
Borja-Aburto	1997	Mexico	Mexico City	94.0	0.02	(-0.02, 0.05)
Hong	1999	South Korea	Inchon	15.4	0.01	(-0.90, 0.92)
Bremner	1999	UK	London	16.0	-0.07	(-0.16, 0.01)
Cadum	1999	Italy	Turin	73.7	0.05	(-0.15, 0.25)
Anderson	2001	UK	West Midlands	24.0	0.04	(-0.10, 0.17)
Saez	2002	Spain	Barcelona	67.5	0.04	(-0.06, 0.14)

Wong	2002	China	Hong Kong	29.3	0.10	(0.04, 0.16)
Le Tertre	2002	France	Le Havre	43.4	-0.20	(-0.67, 0.27)
Le Tertre	2002	France	Lyon	52.0	0.17	(-0.10, 0.44)
Saez	2002	Spain	Madrid	42.1	0.01	(-0.10, 0.12)
Le Tertre	2002	France	Paris	26.0	-0.04	(-0.19, 0.11)
Le Tertre	2002	France	Rouen	57.9	0.21	(-0.21, 0.62)
Le Tertre	2002	France	Strasbourg	37.0	0.00	(-0.24, 0.25)
Le Tertre	2002	France	Toulouse	68.0	0.10	(-0.32, 0.52)
Saez	2002	Spain	Valencia	45.5	0.18	(-0.39, 0.76)
Penttinen	2004	Finland	Helsinki	50.0	0.32	(0.12, 0.52)
Zhang	2006	China	Shanghai	56.1	0.03	(-0.04, 0.11)
Qian	2007	China	Wuhan	47.2	0.06	(-0.04, 0.17)
			Pooled FE	*0.03*		*(0.01, 0.06)*
			Pooled RE	*0.04*		*(0.01, 0.07)*

Heterogeneity chi-squared = 25.46 (d.f. = 18) p = 0.113

* Mean/median daily concentrations (ug/m3)
~ Percentage change in mean number of deaths per 1 ug/m3 increase in O3
$ 95% confidence limit

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IV. American Cancer Society Cancer Prevention II Study (CPS II)

Cohort Information

Enrollment of the American Cancer Society CPS cohort occurred in 1982-1983 and was restricted to persons who were at least 30 years of age in households with at least one individual 45 years of age or older. After supplying written informed consent, subjects completed a questionnaire that included demographic characteristics, smoking history, alcohol use, diet and education[1]. Mortality was ascertained by examination of death certificates in 1984, 1986, and 1988 and subsequently using the National Death Index[2]. The study population included only those participants in CPS-II who resided in US Metropolitan Statistics Areas (MSAs) within the 48 contiguous states or the District of Columbia (based on their address at the time of enrollment), and for whom pollution data were available based on the presence of at least one pollution monitor for $PM_{2.5}$, sulfate, elemental carbon (EC) or ozone within their MSA.

Confounding variables

These variables included individual risk factors collected in the CPS-II questionnaire such as smoking habits and occupational status (see Jerrett et al. (2009)[3] for a complete listing of the variables). Seven ecological covariates obtained from the 1980 US Census (median household income, proportion of the population <125% of the poverty line, percentage of unemployed persons over the age of 16 years, percentage of adults with education less than grade 12, percentage of homes with air conditioning, the Gini coefficient of income inequality and the percentage of the population that is white) were also included in the models. These variables were included at two levels: (1) as the MSA average and (2) as the zip code scale of residence for each subject, deviated from the MSA average (see Krewski et al. (2009)[2] for complete description of the variables).

Exposure Estimates for the American Cancer Society National Analysis

EC estimates were based on the Health Effects Institute (HEI) Air Quality Database (https://hei.aer.com/), which was accessed on June 18, 2009. Annual average values were already calculated based on accepted completeness criteria for the United States Environmental Protection Agency Speciation Trends Network. The completeness of data prior to 2000 limited the number of sites, and as a result we only extracted means for the years 2000-2007. Only sites with an HEI completeness value of 0.5 or above were included. After being joined to the metropolitan statistical areas (MSAs) or the primary metropolitan statistical areas (PMSA) through the longitude and latitude of the site, all monitoring sites within a given MSA/PMSA were averaged. Maximal spatial coverage was available for the years 2003-2006. A three-year average of the annual means was then calculated to assign exposure representing the long-term average concentration of EC in each MSA. In total we were able to join 66 MSAs with ACS health data and data on the other pollutants ($PM_{2.5}$, ozone, and sulfate). Details on the other pollutants and how they were calculated is available in Krewski et al. 2009[4], Jerrett et al. 2009[3], and Krewski et al. 2000[5]. These 66 MSAs formed the basis for our analysis presented in the main paper.

Statistical Modeling Details

We used Random Effects Cox (REC) models to estimate all the mortality risk estimates used in the American Cancer Society Cancer Prevention II National Analysis. The REC models were estimated based on custom software that we developed as part of an HEI project. Full details of the statistical model development and software are available in Krewski et al. (2009)[4] and the software is also available publicly from HEI.

This model is expressed mathematically in the form:

$$h_{ik,s}(t) = h_{0s}(t)\, \eta_k * \exp(\beta' x_{ik})$$

where $h_{ik,s}(t)$ is the hazard function or instantaneous probability of death, at time t for the i^{th} subject in the k^{th} MSA in stratum s, conditional on the random effects η_k. Here, $h_{0s}(t)$ is the baseline hazard function for stratum s, defined by age-gender-race categories. The η_k are positive random variables representing the unexplained variation in the response between MSAs. Only the moments of the random effects need to be specified within our modeling framework:

$E(\eta_j)= 1$, $Var(\eta_k)=\tau^2$.

We assume that given the MSA random effects, responses between subjects are independent. Each model includes 20 individual variables expressed with 44 terms (for smoking, occupational exposures and the like) and the ecological variables as specified above.

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